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by Norman C. Wenger and Jerry Smetana

Lewis Research Center

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LIQUID-HYDROGEN DENSITY MEASUREMENTS USING AN OPEN-ENDED MICROWAVE CAVITY

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SUMMARY

The use of an open-ended microwave cavity to determine the density of liquid hydrogen is discussed. Design curves for the cavity are presented. An experiment that measured hydrogen density by using the cavity is described, and the experimental results obtained are presented and discussed. The problem of measuring hydrogen density when two phases are present is also treated.

INTRODUCTION

One of the major parameters of interest in systems using liquid hydrogen is density. The two most frequent requirements for a density measurement are in the determination of mass and mass flow rates. For example, a common technique for determining the mass of hydrogen in a tank is to measure both the liquid level and the density. The mass flow rate of hydrogen in a line can be determined by measuring both the volumetric flow rate and the density.

A density measurement under flow conditions is generally more difficult than a density measurement in a tank. The density meter for the flow case should have a rapid response and a wide range in order to cover both liquid and gaseous hydrogen, and it should also present a minimal obstruction to the flow. A device that possesses these desirable features is an open-ended microwave cavity.

Microwave cavities have been used for many years to measure the densities of gases. The shift in the resonant frequency when a gas is introduced into an evacuated microwave cavity gives a measure of the dielectric constant of the gas. The density of the gas can then be determined from the dielectric constant data by using either a suitable theory on electric polarization for that particular gas or an empirical or experimentally obtained formula.

To make measurements of the density under flow conditions, it is necessary to make the microwave cavity open-ended. The early work in the field of open-ended cavities was prompted by the need for a device that could be attached to an aircraft or a weather balloon to obtain data on the dielectric constant of the atmosphere as a function of altitude.

An open-ended microwave cavity was recently developed at the Lewis Research Center to determine the feasibility of using such a device for measuring the density of liquid hydrogen. This report presents some experimental data on hydrogen density measurements obtained by using this cavity. The measurements were taken under static conditions.

The first section of this report is devoted to a discussion of some presently available theories that relate the density of hydrogen to its dielectric constant. Design curves for the cavity are then presented followed by a description of the experiment and a discussion of the resulting data. Finally, consideration is given to the problem of density measurements for two-phase hydrogen.

SYMBOLS

a	radius of cavity partition
a_0	molecular radius of hydrogen
b	radius of cavity
c	velocity of light
f	resonant frequency when cavity is filled with dielectric
f_0	resonant frequency when cavity is evacuated
k_0	free space wave number, $2\pi f_0/c$
K	relative dielectric constant
K_{eff}	effective relative dielectric constant of mixture
K_g	relative dielectric constant of gas
K_l	relative dielectric constant of liquid
L	distance between partitions in cavity
m	mass of hydrogen molecule
Q	2π times ratio of energy stored in cavity to energy dissipated per cycle of oscillation
r_0	radius of gas bubble

- α average polarizability of hydrogen molecule
- δ fraction of volume occupied by gas
- ϵ_0 permittivity of free space
- λ wavelength of electromagnetic waves
- ρ mass density of hydrogen

DIELECTRIC CONSTANT OF HYDROGEN

In practice, the Clausius-Mossotti equation is normally used to relate the density of hydrogen to its relative dielectric constant. The standard form of this equation is

$$\rho = \frac{3\epsilon_0 m}{\alpha} \frac{K - 1}{K + 2} \tag{1}$$

where ρ is the mass density of hydrogen, K the relative dielectric constant, m the mass of a hydrogen molecule, α the average polarizability of a hydrogen molecule, and ϵ_0 the electric permittivity of free space.

The Clausius-Mossotti equation is a derived equation that is based on the assumption that each molecule "sees" an internal field given by the Lorentz local field formula (ref. 1, p. 177). Böttcher has shown that the Lorentz formula gives the average internal field over the whole dielectric rather than the internal field that each molecule experiences (ref. 1, pp. 177 to 181). Thus, the Clausius-Mossotti equation is in error. This error may become large, particularly for high densities.

A more accurate formula for computing density from relative dielectric constants has been derived by Böttcher (ref. 1, pp. 205 to 212). Böttcher's formula is based on a model for computing the internal field that pictures the molecule under study to consist of a polarizable point dipole centered in a spherical void. The radius of the void a_0 corresponds to the molecular radius. The remaining molecules are assumed to act like a homogeneous dielectric medium. The result of this derivation is formulated in Böttcher's equation

$$\rho = \frac{m\epsilon_0}{3\alpha} \frac{(K - 1)(2K + 1)}{K} - \frac{m}{6\pi a_0^3} \frac{(K - 1)^2}{K} \tag{2}$$

It is interesting to note that if the molecular radius a_0 satisfies the equation

$$\frac{4\pi a_0^3}{3} \frac{\rho}{m} = 1$$

which is equivalent to requiring the molecular radius to be of such a value that the molecules fill all available space, then Böttcher's equation and the Clausius-Mossotti equation are equivalent.

A slight rearrangement of Böttcher's equation reveals that a plot of $3K\rho/[(K-1)(2K+1)]$ against $(K-1)/(2K+1)$ will be a straight line of slope $-m/2\pi a_0^3$ and intercept $m\epsilon_0/\alpha$. Such a plot using a representative sample of Stewart's data (ref. 2) is shown in figure 1. These experimental data consist of 205 points of dielectric constant as a function of density for para-hydrogen over the density range from 2 to 80 kilograms per cubic meter. The error in these data is estimated to be less than 0.1 percent. The straight line shown in figure 1 is a least-square-error fit to all 205 data points. The line has a slope of -53.92 kilograms per cubic meter and an intercept of 332.1 kilograms per cubic meter. The deviation between the data points and the straight line is everywhere less than 0.1 percent. Since this deviation never exceeds the error in Stewart's data, it can be concluded that Böttcher's equation relates the density of para-hydrogen to its dielectric constant with an error less than 0.1 percent over the range of densities from 2 to 80 kilograms per cubic meter.

A major assumption in the derivation of Böttcher's equation is that the fields are either static or are varying so slowly with time that dynamic effects and damping mech-

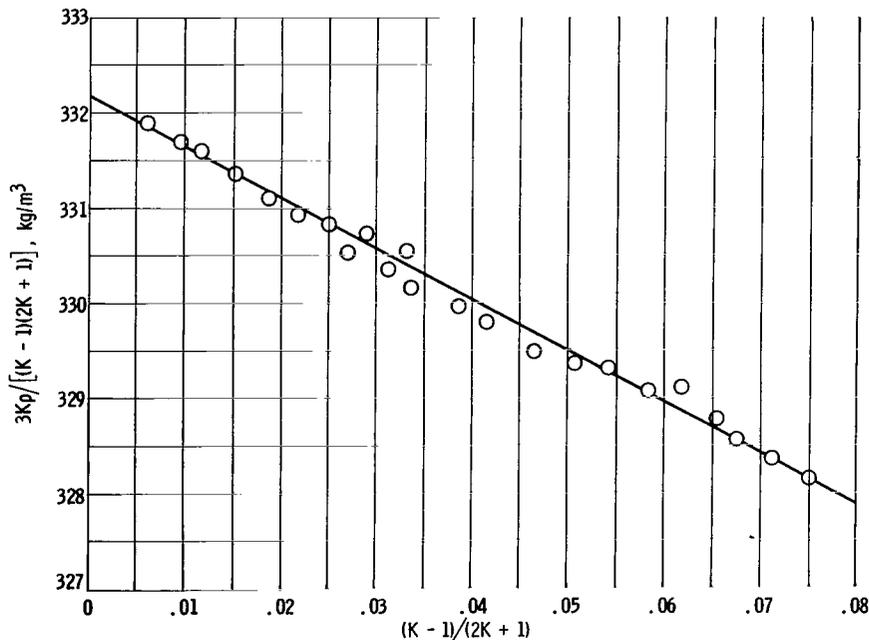


Figure 1. - Least-square-error fit to Stewart's data (ref. 2).

anisms within the molecules will not appreciably affect the dielectric constant. If the fields vary rapidly with time, these effects will cause the dielectric constant to differ from its low-frequency value and will make the dielectric slightly conductive. This last effect is one of the major factors in determining whether it is feasible to use a microwave cavity to determine density, since damping mechanisms at the cavity resonant frequency may obscure the resonance.

OPEN-ENDED CAVITY

The open-ended cavity used in the experiment is shown in figure 2. A theoretical analysis of microwave cavities of this type is covered in reference 3. This section presents results of the analysis in the form of design curves that show the relation between

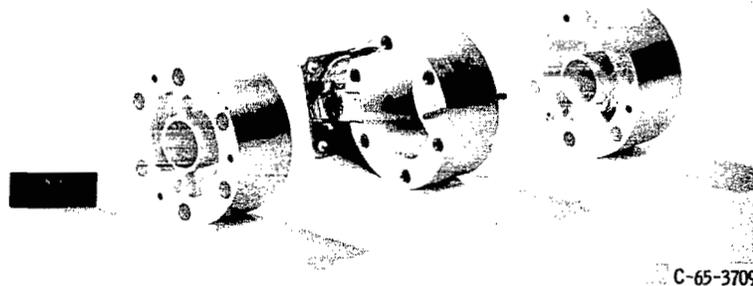


Figure 2. - Open-ended microwave cavity.

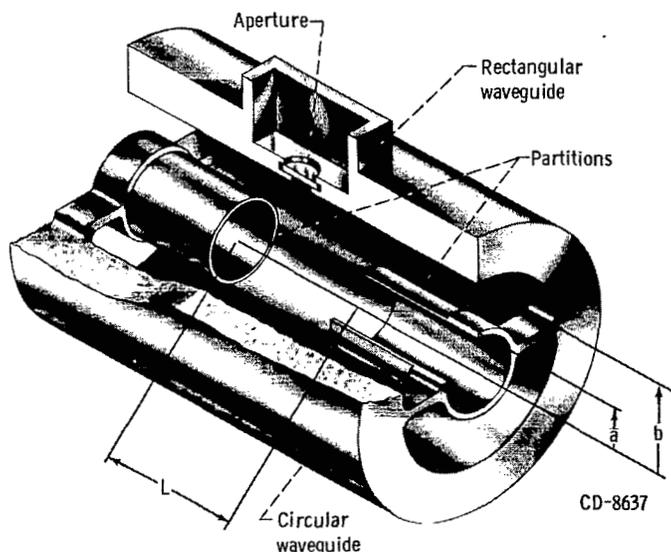


Figure 3. - Model of open-ended cavity.

the resonant frequency of the cavity and the cavity dimensions and relates the resonant frequency to the relative dielectric constant of the material in the cavity.

A diagram of the cavity is shown in figure 3. It consists of a circular waveguide of radius b that is terminated at each end with thin coaxial cylindrical partitions of radius a that are separated by a distance L . Microwave energy is supplied to the cavity through the small aperture in the side wall. If the cylindrical partitions are sufficiently long (ref. 3),

they will act as perfect reflectors to the microwave energy over a frequency range whose limits are dependent on the dimensions a and b . The cavity is resonant when the spacing between the partitions is adjusted so that the reflections are constructive. This condition occurs when the spacing L is approximately an integral multiple of one-half wavelength.

The required spacing for the TE_{011} mode of oscillation is shown in figure 4 in the form of curves of L/b against the dimensionless resonant frequency $k_0 b$ for typical values of the parameter b/a . The free-space wave number k_0 is equal to $2\pi f_0/c$, where f_0 is the resonant frequency of the cavity when it is evacuated, and c is the velocity of light. The curves in figure 4 have been obtained by assuming that the cavity walls are perfectly conducting and that the partitions are infinitely thin. The TE_{011} mode of oscillation was selected because of its high Q .

The dimensionless resonant frequency $k_0 b$ is bounded from below by 3.832 for all values of b/a . The upper limit of $k_0 b$ is, however, a strong function of b/a . The fact that $k_0 b$ is bounded implies that the range of resonant frequencies for a fixed value of b

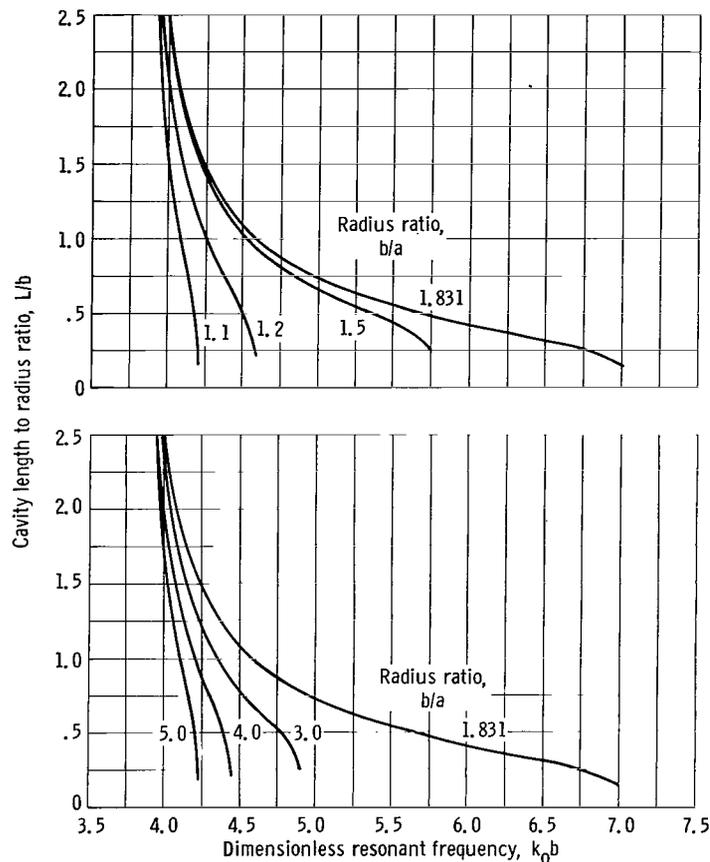


Figure 4. - Ratio of cavity length to radius against dimensionless resonant frequency for constant values of radius ratio.

is also bounded. For example, if the cavity is to be used in a 0.05-meter inside-diameter flow line, the dimension b would be 0.025 meter. Since f_0 is in the range $3.832(c/2\pi b) < f_0 < 7.015(c/2\pi b)$ for $b/a = 1.831$, the resonant frequency has a lower limit of 7.319 gigahertz and an upper limit of 13.40 gigahertz. If b/a differs greatly from 1.831, the upper limit is, of course, reduced considerably. The cavity can be made to resonate at any frequency between these limits by selecting the proper spacing L as determined from figure 4.

The cavity shown in figure 2 was constructed of silver-plated brass with $b/a = 2.082$. This value of b/a corresponds to locating the cylindrical partition at the radius of maximum electric field intensity for the TE_{01} circular waveguide mode. The dimensions of the cavity are as follows: $b = 0.02060$ meter, $a = 0.00989$ meter, and $L = 0.02555$ meter. The value for a is taken as the mean radius of the partition, which has a wall thickness of 0.0005 meter.

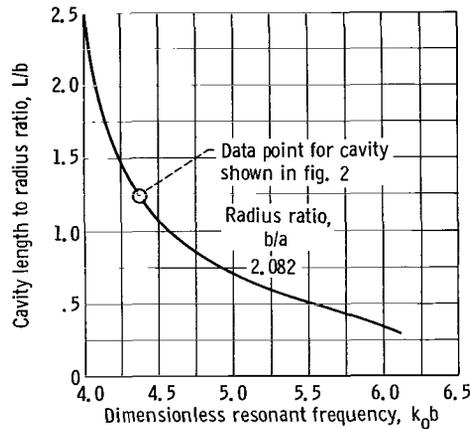


Figure 5. - Ratio of cavity length to radius against dimensionless resonant frequency for radius ratio of 2.082.

A curve of L/b against $k_0 b$ for $b/a = 2.082$ is shown in figure 5. The numerical values for b , a , and L predict that $k_0 b$ should be 4.376, which corresponds to a resonant frequency of 10.144 gigahertz. The measured value for the resonant frequency was 10.183 gigahertz. The 0.4 percent difference between the theoretical and experimental resonant frequencies is probably a result of the finite wall thickness of the partitions and the presence of the aperture. These effects were not considered in the theory.

The resonant frequencies predicted by the curves in figures 4 and 5 are for the case where the cavity is evacuated. It can easily be shown that if the cavity is completely filled with a homogeneous, isotropic, dielectric material of relative dielectric constant K , the resonant frequency will be reduced by the factor $1/\sqrt{K}$. Thus, if f is the resonant frequency when the cavity is filled with a dielectric and if f_0 is the resonant frequency when it is evacuated, K is simply

$$K = \left(\frac{f_0}{f}\right)^2 \quad (3)$$

Equation (3) is the basic equation that will be used to relate the relative dielectric constant of hydrogen to measured resonant frequencies.

Care must be taken in selecting the resonant frequency f_0 so that both f_0 and f are within the bandwidth of available instrumentation. For example, if X-band instru-

mentation is used, the available frequency range is 8.2 to 12.4 gigahertz. If f_0 is set equal to the upper limit of 12.4 gigahertz, a dielectric material in the cavity with a relative dielectric constant of 2.28 will reduce the resonant frequency to the lower limit of 8.2 gigahertz. Thus, if both f_0 and f are to be in X-band, the relative dielectric constant must be less than 2.28. This same result also holds for S-band, K-band, etc., since the ratio of the upper frequency limit to the lower frequency limit for these bands is approximately 1.5. Since hydrogen has a relative dielectric constant in the range 1.0 to 1.3, no difficulty is encountered in keeping both f_0 and f within the band if f_0 is properly selected.

EXPERIMENT

The experiment to measure the density of liquid hydrogen by using an open-ended cavity was performed in a stainless-steel vacuum-jacketed tank approximately 0.75 meter in diameter and 2 meters high. A stainless-steel, vacuum-jacketed pressure vessel approximately 0.25 meter in diameter and 0.25 meter high was suspended near the bottom of this tank. A diagram of the pressure vessel is shown in figure 6.

The open-ended microwave cavity was oriented in the pressure vessel with the open ends at top and bottom to eliminate the possibility of collecting gas bubbles within the

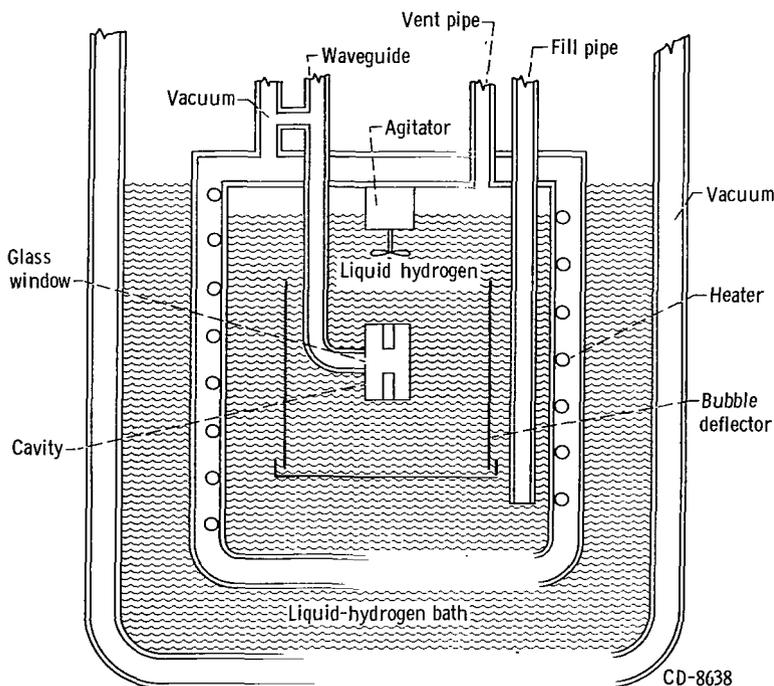


Figure 6. - Pressure vessel.

cavity. An agitator was provided to ensure that stratification of the liquid did not occur, and a bubble deflector was placed around the cavity to divert any bubbles that may have been generated along the bottom of the pressure vessel.

The aperture in the side wall of the cavity was sealed with a glass window to prevent hydrogen from entering the waveguide. The window was constructed by pouring molten glass into a metal ring that had an inside diameter of 0.080 meter and a thickness of 0.025 meter. The ring was then soldered into the aperture of the cavity. The window was tested for leaks by cold-shocking with liquid nitrogen and then pressure-testing with oil at 150 atmospheres and gaseous helium at 15 atmospheres. No leaks were detected.

The experiment was conducted with the large tank filled with liquid hydrogen vented to the atmosphere. This provided a bath around the pressure vessel at a temperature of 20.2° K. The pressure and temperature of the hydrogen in the pressure vessel could be varied by forcing gaseous hydrogen from a gas storage cylinder into the fill pipe of the vessel or by manually adjusting the current in a heating element that was wrapped around the inner tank of the vessel. In this way, pressures in the range from 1 to 7 atmospheres and temperatures in the range from 21° to 27° K were achieved to give a density range of 57 to 72 kilograms per cubic meter.

The density of the hydrogen in the vessel was determined by using the microwave cavity to measure the dielectric constant and then computing the density from Böttcher's equation, and by measuring the pressure and temperature of the hydrogen and then using an equation of state for para-hydrogen developed by the National Bureau of Standards (ref. 4) to compute the density. Before presenting the results of these density measurements, the basic pressure, temperature, and resonant frequency measurements will be discussed in some detail.

Measurements

Pressure. - The pressure of the hydrogen was measured with an absolute pressure transducer located on the fill pipe of the pressure vessel external to the tank. The transducer was calibrated immediately before the start of the experiment at intervals of 0.5 atmosphere. The error in the pressure measurement was estimated to be less than 0.05 atmosphere.

The pressure of the hydrogen at the location of the cavity is somewhat higher than that indicated by the pressure transducer since the cavity is approximately 2 meters lower in the liquid. However, since this difference was computed and was less than 0.02 atmosphere, it was neglected.

Temperature. - The temperature of the hydrogen was determined by using five platinum-resistance temperature sensors. Two of the sensors were mounted in direct

contact with the cavity body. The remaining sensors were suspended with nylon thread in the pressure vessel at various levels. The purpose of these sensors was to detect the presence of temperature gradients within the vessel and to compare the cavity temperature with the liquid temperature.

The platinum sensors were connected electrically in series with a standard resistor and a constant current source. The resistance of each sensor could be determined by measuring the voltage drop across the sensor and across the standard resistor with a digital voltmeter and then by computing the product of the standard resistance and the ratio of these two voltages. In practice, this computation was eliminated by adjusting the current source so that the voltage drop across the standard resistance in millivolts was equal to the resistance of the standard in ohms. In this way, the voltage drop across each sensor in millivolts was equal numerically to its resistance in ohms. Since the digital voltmeter was used merely as a transfer device, it contributed little to the error of the resistance measurement. It was estimated that the error in the measurement of the sensor resistances was less than 0.05 percent.

Each of the sensors was calibrated against a standard platinum sensor that was calibrated by NBS. The calibration error for the sensors was less than 0.03° K. Since the temperature coefficient of these sensors was approximately 12 percent per K° over the range of temperatures used in the experiment, the error in the sensor resistance of 0.05 percent corresponded to a temperature error of 0.004° K. Thus, the total error in the temperature measurement was approximately the calibration error of 0.03° K.

Density. - Over the range of temperatures and pressures encountered in the experiment, the density of liquid hydrogen changed approximately 1.5 percent per $^{\circ}$ K at constant pressure and approximately 0.2 percent per atmosphere at constant temperature. Thus, the error in determining the density was 0.05 percent from the temperature error, plus 0.01 percent from the pressure error, plus the error in the equation of state of 0.02 percent (ref. 4, p. 10) for a total probable error of 0.06 percent.

Resonant frequency. - A block diagram of the instrumentation for measuring the resonant frequency of the open-ended cavity is shown in figure 7. The sweep oscillator, reference cavity, and dual-beam oscilloscope were located in a control room approximately 30 meters from the experiment. The sweep oscillator was adjusted to a sweep over a 1-megahertz range every 0.01 second with the center frequency of this range being adjustable from 8.2 to 12.4 gigahertz. The reflected signal from the open-ended cavity was detected and observed on one beam of the oscilloscope, and the transmitted signal from the reference cavity was detected and observed on the second beam.

The resonant frequency measurement was carried out by manually adjusting the center frequency of the sweep oscillator until the response curve of the open-ended cavity was observed on the oscilloscope. The reference cavity was then tuned so that the peak of the response curve for the reference cavity aligned with the peak of the response curve

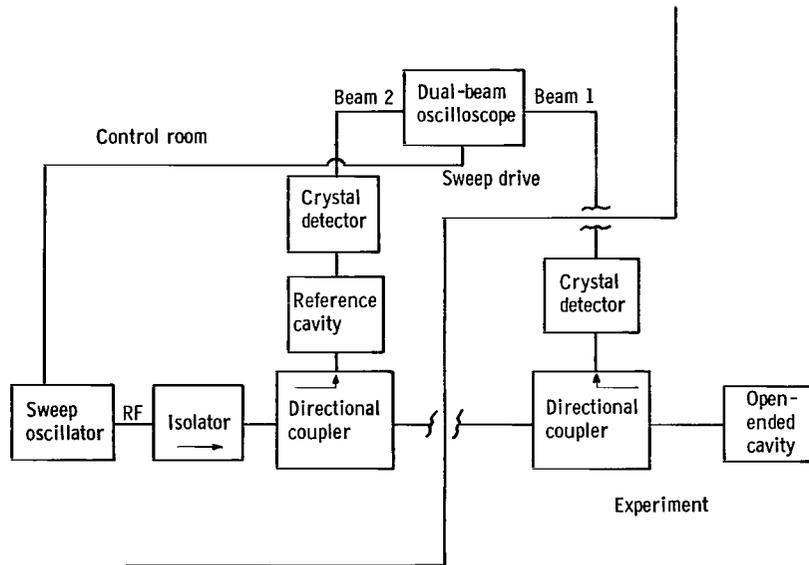


Figure 7. - Microwave instrumentation.

of the open-ended cavity. The dial reading on the reference cavity then corresponded to the resonant frequency of the open-ended cavity. The reference cavity was calibrated against a frequency standard after the completion of the experiment. The estimated error in the frequency measurements was less than 0.01 percent.

Experimental Procedure

The initial portion of the experiment consisted of cooling the pressure vessel to liquid-hydrogen temperature with the vessel evacuated to obtain f_0 , the resonant frequency of the cavity when it is evacuated. The resonant frequency of the cavity increased 0.39 percent as it cooled from its room temperature value of 10.183 gigahertz to 10.224 gigahertz at 24.5° K. This percentage increase is in exact agreement with the coefficient of thermal expansion for brass for this temperature change (ref. 5).

The Q of the cavity which is 10^4 at room temperature increased to approximately 10^5 at liquid-hydrogen temperature. This increase was expected since the cavity Q is proportional to the square root of the conductivity of the cavity surface material, in this case silver, which increases by a factor of approximately 10^2 for this temperature change.

After f_0 was determined, liquid hydrogen was admitted into the vessel, and resonant frequency measurements were taken at various densities. The measurements were only taken when all temperature sensors indicated the same temperature. The cavity Q remained at its previous value of 10^5 for all values of density covered. The fact that the

the cavity Q did not decrease indicates that the electrical losses of liquid hydrogen at these frequencies is extremely small. This result was significant since the cavity technique for measuring hydrogen density would not be feasible if the electrical losses of hydrogen were high. If the electrical losses were appreciable, the resonant frequency could not be determined accurately as a result of the broadening of the cavity-response curve.

Results

The relative dielectric constant of hydrogen was computed for each resonant frequency data point by using equation (3). The value used for f_0 was its value at 24.5°K of 10.224 gigahertz. The variation of f_0 with temperature over the range 21° to 27°K was negligible. It was estimated that the probable error in determining the relative dielectric constant was less than 0.03 percent since the dielectric constant is given by the square of the ratio of two frequencies that are each known to within 0.01 percent.

The density of hydrogen was computed from the dielectric constant data by using Böttcher's equation. It can easily be shown by computing $\partial\rho/\partial K$ from Böttcher's equation that the percent error in density is approximately five times the percent error in dielectric constant, or 0.15 percent.

The results of the experiment are presented in figure 8 in the form of data points showing the difference in percent between the density measurement taken with the cavity and the density measurement taken from the pressure-temperature data using the NBS equation of state. As shown, the percentage difference between the two techniques is less than 0.07 percent which is well within the expected experimental error of the cavity measurement. The results demonstrate that Böttcher's equation will apply for hydrogen when the relative dielectric constant is measured at microwave frequencies.

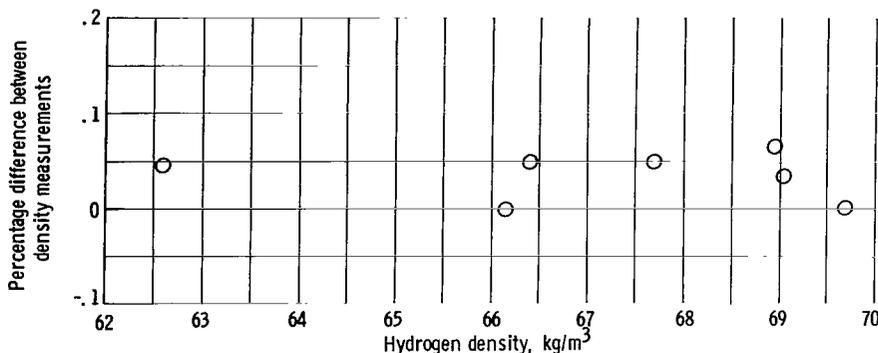


Figure 8. - Percentage difference between hydrogen density determined from cavity and from pressure-temperature data against density.

TWO-PHASE HYDROGEN

In many cases where hydrogen density measurements are required, such as in mass flow rate measurements, for example, hydrogen may exist in two phases. Equations (2) and (3) relate the resonant frequency of the cavity to the density of the hydrogen only for the case where the hydrogen is homogeneous. If the hydrogen is nonhomogeneous, as would be the case if bubbles were present, the cavity will still resonate, but the resonant frequency may no longer be indicative of the density.

It can be shown (ref. 6) that the effective relative dielectric constant of a mixture K_{eff} that consists of liquid hydrogen with relative dielectric constant K_{ℓ} and gaseous hydrogen with relative dielectric constant K_g is given by

$$K_{\text{eff}} = K_{\ell} - 3\delta K_{\text{eff}} \frac{K_{\ell} - K_g}{2K_{\text{eff}} + K_g} \quad (4)$$

where δ is the fraction of the total volume that is occupied by the gas. Equation (4) is a derived equation that is based on the assumption that the fraction of the total volume occupied by the gas is very small (i. e., $\delta^2 \ll 1$) and that the gas bubbles are uniformly distributed in the liquid and are spherical with a radius r_0 that is much less than the wavelength λ at which K_{eff} is measured.

An "effective" density for the two-phase mixture can be computed by using equation (4) and Böttcher's equation. To determine the difference between this effective density and the actual density, the case where $K_{\ell} = 1.20$ and $K_g = 1.00$ is considered. Figure 9 shows a plot of the difference in percent between the effective density and the actual density against δ . As shown, this difference is less than 0.1 percent for values of δ up to 0.45. Thus, it can be concluded that an open-ended cavity can be used to make accurate two-phase hydrogen density measurements if the gas bubbles are sufficiently small and uniformly distributed so that the assumptions on which equation (4) is based are not violated.

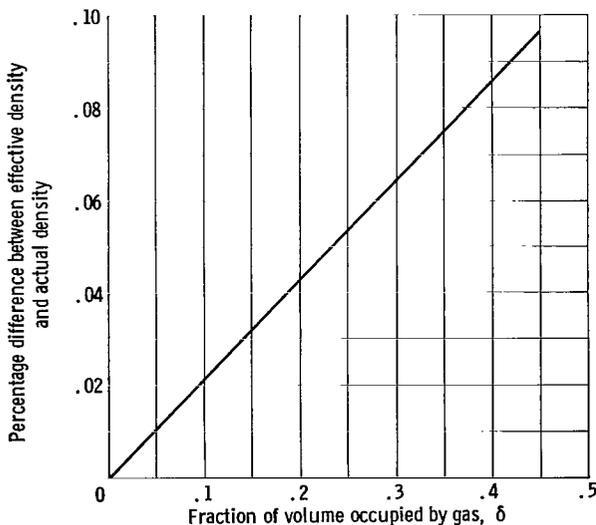


Figure 9. - Theoretical percentage difference between effective density and actual density against fraction of volume occupied by gas. Relative dielectric constant of liquid, 1.20; relative dielectric constant of gas, 1.00.

An experiment was performed to

determine how small the bubbles must be before equation (4) applied and to determine the error for the case of larger bubbles. The hydrogen was simulated by a polystyrene-based plastic-foam material with a nominal relative dielectric constant of 1.2. The cavity used in this experiment was approximately the same size as the one shown in figure 2 (p. 5) except that it had solid end walls since it was not necessary to have material flow through the cavity.

The cavity was first filled with a cylindrical sample of this plastic foam that fitted snugly in the cavity. Measuring the resonant frequency of the cavity gave the relative dielectric constant of the sample as 1.1998 ± 0.0004 . This value of dielectric constant corresponds to a hydrogen density of 62.13 kilograms per cubic meter.

This sample was then cut into cubes approximately 0.00625 meter on a side. A portion of these cubes was placed in the cavity so that they filled the cavity from end to end, leaving only small gaps between the cubes. These gaps simulated gaseous hydrogen bubbles, and the foam material simulated the liquid hydrogen. The ratio of the radius of the simulated bubbles to the wavelength at the resonant frequency was of the order of 0.1. The ratio of the simulated gas volume to the total volume δ could be changed by varying the packing density of the cubes. The exact value of δ was calculated from the ratio of the weight of the cubes to the weight of the initial sample of foam.

Resonant frequency measurements were made for several arrangements of the cubes within the cavity so that the effect of varying the location of the simulated gas bubbles could be observed. The results are presented in figure 10 and show the difference in percent between the effective density and the simulated density as a function of δ . The length of the vertical lines indicates the range over which this difference varied by changing the locations of the simulated gas bubbles. The results indicate that the simulated bubbles were much too large to satisfy the assumptions on which equation (4) is based.

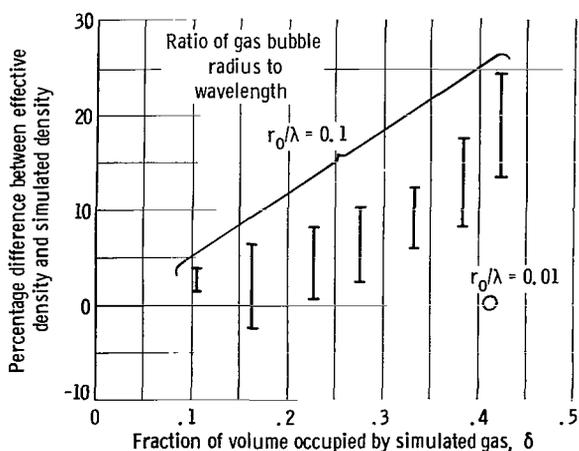


Figure 10. - Measured percentage difference between effective density and simulated density against fraction of volume occupied by simulated gas.

In addition, there is considerable error due to varying the locations of the simulated bubbles.

The experiment was repeated with the cavity filled with small particles of the foam material that had a maximum dimension of 0.0007 meter, which gave a simulated gas bubble radius to wavelength ratio of the order of 0.01. Since the particles were so small, the packing density could not be easily changed. For the case where $\delta = 0.413$, the difference between the effective density and the simulated density was only 0.22 percent, which is much closer to the predicted

value of 0.09 percent (see fig. 9). Therefore, it can be concluded that the cavity technique can be used for measuring the density of two-phase hydrogen with an error less than 1 percent if the ratio of the bubble radius to the operating wavelength can be made of the order of 0.01 or less and if the bubbles can be uniformly distributed in the liquid.

CONCLUSIONS

Hydrogen density measurements that were made by using an open-ended microwave cavity were in excellent agreement with density calculations that were based on pressure-temperature data. Accurate density measurements could be taken for two-phase hydrogen if the hydrogen gas bubbles could be reduced sufficiently in size and distributed uniformly in the liquid.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, July 28, 1966,
128-31-06-77-22.

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